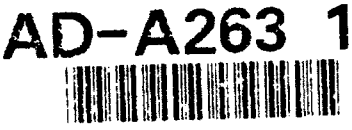

ARMY RESEARCH LABORATORY

Tomographic Analysis of Line-of-Sight Infrared Spectra of Low-Pressure Flames

Kevin L. McNesby
Robert A. Fifer

ARL-TR-108 April 1993

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13. ABSTRACT *(Maximum 200 words)*

Line-of-sight Fourier transform infrared absorption spectra through a 17-torr methane/nitrous oxide flame have been tomographically analyzed using Abel's equations. Relative absorbances for N_2O, CH_4, CO_2, NO, and CO have been mapped for various "slices" of the flame at different heights in the flame. Results indicate that the region of maximum NO and CO formation occurs prior to the luminous region of the flame. The technique is shown to be useful in obtaining data from systems for which line of sight data may be unusable.

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1. INTRODUCTION

Several techniques have been applied to the study of low-pressure flames (Penner, Wang, and Bahadori 1984; Limbaugh 1985). One of the least used is Fourier transform infrared spectroscopy (FTIR), even though a major advantage of using FTIR spectroscopy is that the technique is non-perturbing to the system under investigation (Thorne and Melius 1990). However, drawbacks inherent to FTIR experiments on combusting systems such as line-of-sight limitations (McNesby and Fifer 1991), lack of sensitivity, difficulty in quantifying species and temperature (Anderson and Griffiths 1977), detector nonlinearity (Solomon et al. 1986), and limitations imposed by spectrometer resolution and apodization (Anderson and Griffiths 1975) (if any) have discouraged many investigators. It is not surprising that mass spectrometry, even though it is an intrusive technique, has become a more widely used tool for flame diagnostics (Howard et al. 1992).

The most ubiquitous of the limitations for FTIR transmission spectroscopy of flames is that it is a line-of-sight technique. Recently, Best et al. (1991) published a paper reporting the first use of tomography coupled with FTIR spectroscopy for reconstruction of localized spectra of an axisymmetric flame from line-of-sight absorption measurements. Previously, nonmedical applications of computed tomography have been mainly single wavelength studies in emission and absorption of reacting flows (Emmerman et al. 1980; Best et al. 1991; Hughey and Santavicca 1982).

Although the body of literature on the subject is extensive (Barrett and Swindell 1981; Cormack 1963; Dasch 1992; Penner, Wang, and Bahadori 1984; Limbaugh 1985), tomographic analysis has not become a standard analytical tool in chemistry. In this report, we recount our own efforts at applying tomographic analysis to line-of-sight spectra of low-pressure flames. This work is an extension of our initial efforts reported previously at tomographic reconstruction of FTIR flame spectra (McNesby and Fifer 1992).

2. BACKGROUND

Computed tomography allows the reconstruction of a three-dimensional image of an object by the stacking of two-dimensional "slices" of that object (Shepp and Logan 1974). In computed tomography, obtaining the "slices" is accomplished by computer manipulation of a series of sets of evenly spaced parallel projections through an object. Each set of parallel projections is taken from a different angle or view of the object. When the object is axisymmetric, only one set of parallel projection data is necessary.

Reconstruction of the image of an axisymmetric object from a single set of parallel projections may be accomplished algebraically (the "onion peel" method) using Fourier transforms or by using Abel's equations (Hughey and Santavicca 1982). In the work reported here, Abel's equations are used to analyze FTIR transmission spectra (the raw data input to the reconstruction program are absorbance data) of low-pressure burner flames.

The line-of-sight absorbance at a given frequency for a single absorbing species through an optically thin, axisymmetric medium may be given by

$$g(x) = 2 \int_x^1 f(r)\, r\, dr \,/\, (r^2 - x^2)^{1/2} \qquad (1)$$

where g(x) is the line-of-sight absorbance (projection) through the medium at lateral position x and f(r) is the product of the absorption coefficient times the pressure at radial position r within the axisymmetric medium (Hughey and Santavicca 1982) (see Figure 1). If more than one species absorbed, then f(r) would be a sum of absorption coefficients times pressure. Equation 1 may be solved for f(r) using the Abel transformation

$$f(r) = -1/\pi \int_r^1 g'(x)\, dx \,/\, (x^2 - r^2)^{1/2}$$

where $g'(x)$ denotes the derivative with respect to x of the function g(x). In general, the greater the number of projections, or, in other words, the more parallel line-of-sight spectra obtained, the better the reconstruction (although oversampling may increase error (Dasch [1992]). However, errors in original data tend to be amplified by the transformation process (Hughey and Santavicca 1982), and reconstruction of regions of abrupt change such as at the edge of a flame where large temperature, species, and density gradients occur may be inaccurate (Hughey and Santavicca 1982). Several investigators have tried to determine the best method of overcoming these limitations (Hughey and Santavicca 1982; Dasch 1992).

The general method of solution when Abel's equations are used is to divide the data g(x) into segments and then to fit each of these segments to a polynomial for which an analytical solution to Equation 2 may be obtained (Hughey and Santavicca 1982; Dasch 1992). The process of fitting the data to a function is equivalent to smoothing the data. Smoothing is usually necessary to minimize the noise amplification in the reconstruction mentioned above. Such smoothing or filtering is common to most methods of reconstruction from projection data (Hughey and Santavicca 1982; Dasch 1992).

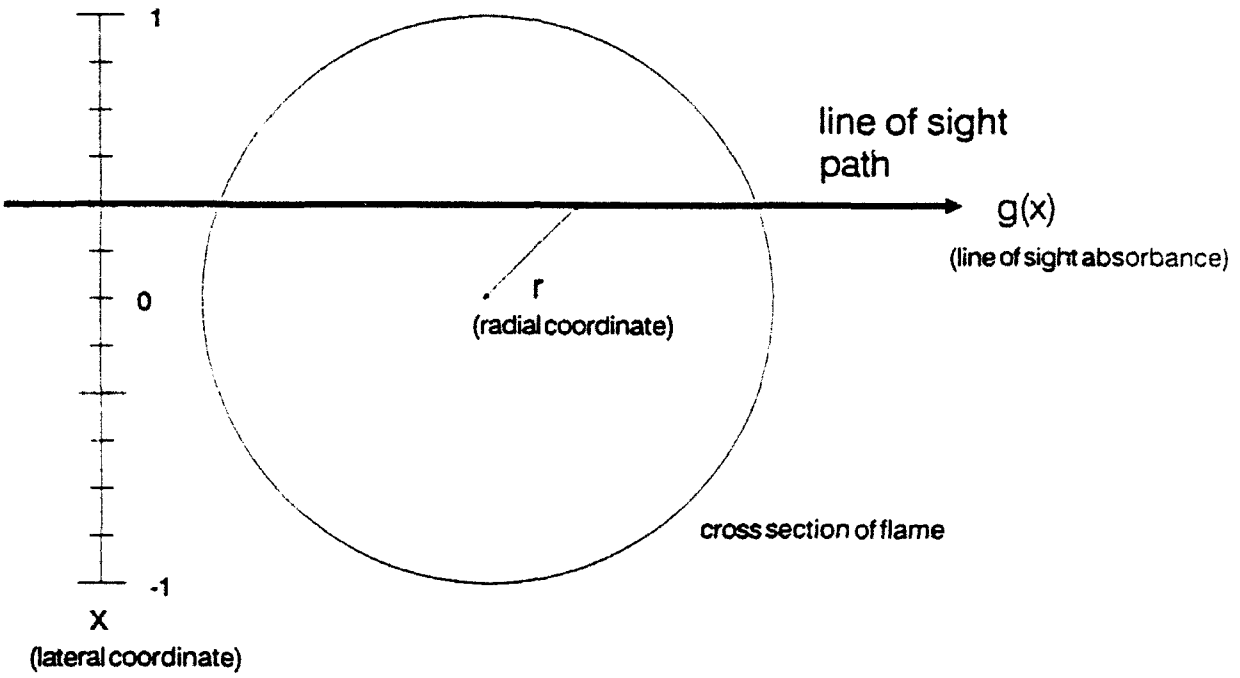

Figure 1. By moving the line-of-sight path to different values of the lateral coordinate, x, different line-of-sight absorbances, g(x), may be obtained.

3. EXPERIMENTAL

A stoichiometric methane/nitrous oxide (CH_4/N_2O) flame was used in all experiments reported here. Total flow rate was 1.8 standard litres per minute. The overall chemical reaction was

$$CH_4 + 4\ N_2O \rightarrow CO_2 + 2\ H_2O + 4\ N_2 \quad .$$

Gases were from Matheson Industries, Inc., and were used without further purification. Pressure within the burner chamber was maintained at 17 (+/-0.2) torr. No shroud gas was used in the experiments reported here. Once the flame had stabilized, the pressure within the chamber slowly increased as the chamber heated up. Over the course of an experiment (several hours), the pressure within the chamber typically rose a few hundred millitorr. The stainless steel frit upon which the flame was supported and through which the premixed gases flowed was 70 mm in diameter. Line-of-sight spectral data were collected for 200 scans at 8-cm^{-1} resolution for each projection on a Mattson Polaris FTIR spectrometer. The spectrometer and beam path external to the burner chamber were purged with dry nitrogen gas to

eliminate absorbance from atmospheric gases. The light beam from the interferometer was directed through a low-pressure housing which contained a McKenna Industries flat flame burner modified for low-pressure operation. Beam size was regulated by 0.75-mm circular apertures on interferometer and Hg-Cd-Te detector sides of the low-pressure housing. Combustion gas flow was controlled by an MKS Inc. Model 147B flow controller. Pressure was controlled using a hand-operated valve in the exhaust line between the vacuum chamber and the Leybold Inc. Model 100 vacuum pump. It was found that for the experiments reported here, hand regulation of the pressure was often better than that achieved using an exhaust line mounted butterfly valve slaved to the MKS capacitance manometer used to monitor pressure. No correction was applied to account for beam divergence or for beam walking in any of the spectra collected (Griffiths and de Haseth 1986). The rest of the experimental apparatus has been described in detail in a previous report (McNesby and Fifer 1992).

Reconstruction was accomplished using a nonderivative solution to Abel's equations employing a least squares fit of spline functions to the experimental data (Deutsch and Beniaminy 1983). For each "slice" of the flame investigated, 20 projections were obtained on one side of the burner axis, with the first projection taken through the burner axis. The beam through the burner flame was 1.3 mm in diameter (95% of the radiant power of the apertured interferometer beam was contained within this diameter). Projections were spatially separated by 2 mm (beam center to beam center). Because of space restrictions within the burner chamber, it was not possible to obtain a complete set of spectra for each side of the burner axis. So, prior to reconstruction, the projections through one side of the burner axis were reflected about the burner axis. A check of parallel projections taken at equal distances from opposite sides of the burner axis (where possible) gave spectra which were superimposable.

4. RESULTS AND DISCUSSION

The appearance of the 17-torr CH_4/N_2O flame used in these experiments is shown in Figure 2. Note the curvature exhibited at the underside of the luminous region of the flame. This curvature indicates that the flame is approaching the "blow off" point (Thorne and Smith 1988), i.e., the pressure and flow rate combination are such that the burner frit will no longer support the flame. Also evident is a nonluminous zone approximately 4 mm in height immediately above the burner surface. Figure 3 shows a series of parallel line-of-sight absorbance spectra (evacuated chamber used as reference), separated by 4 mm, through the flame shown in Figure 2, at a height above the burner (HAB) surface of 0.65 mm. The constant CO_2 absorbance (centered at 2,349 cm^{-1}) due to "cold" gases (i.e., not within the vertical cylinder proscribed by the burner diameter) outside the flame region is evident in each spectrum. Figure 4 shows the tomographic reconstruction using the spectra shown in Figure 3. This figure is dominated by the

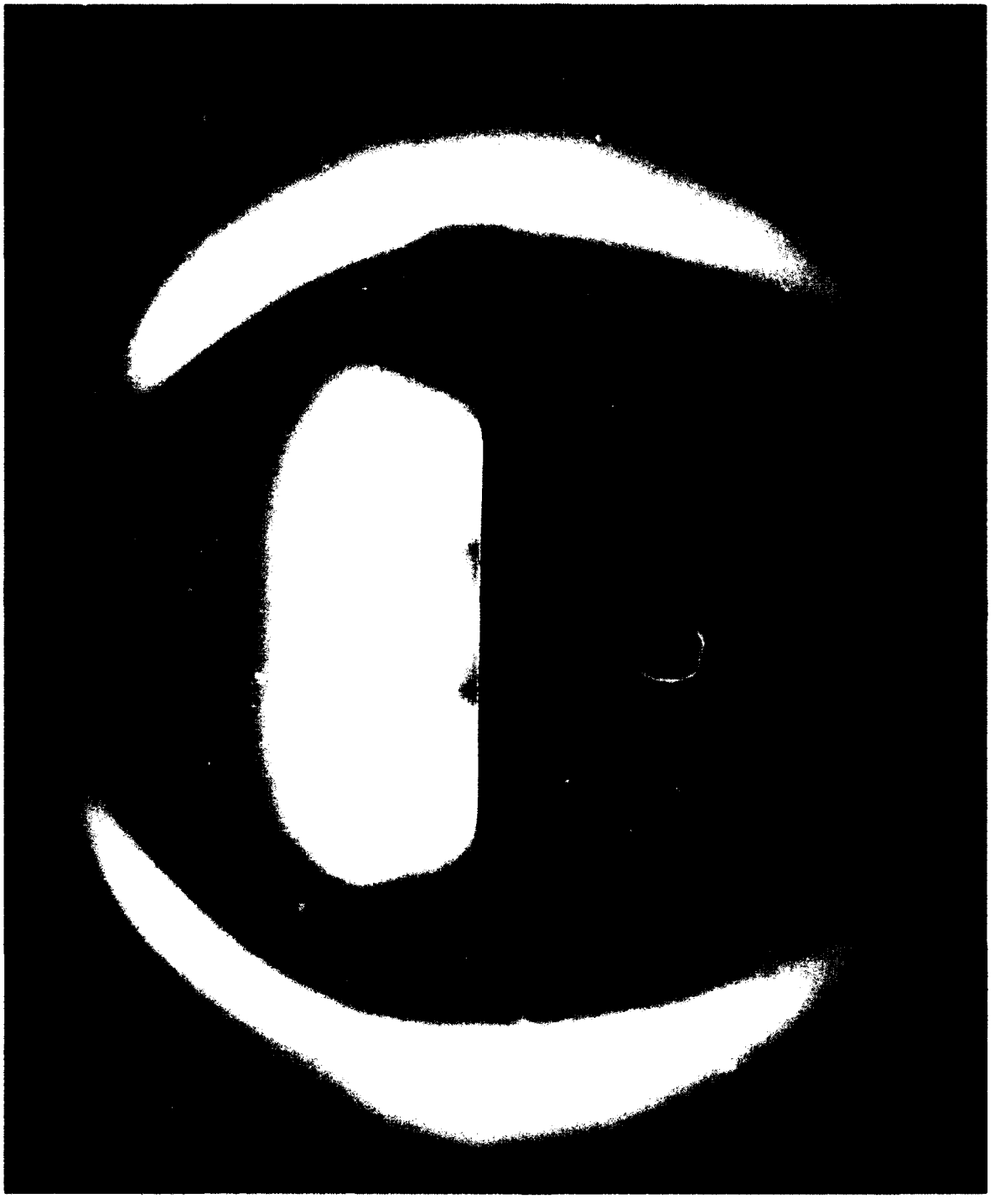

Figure 2. The 17-torr methane/nitrous oxide flame used in these experiments. Note the curvature of the luminous region of the flame.

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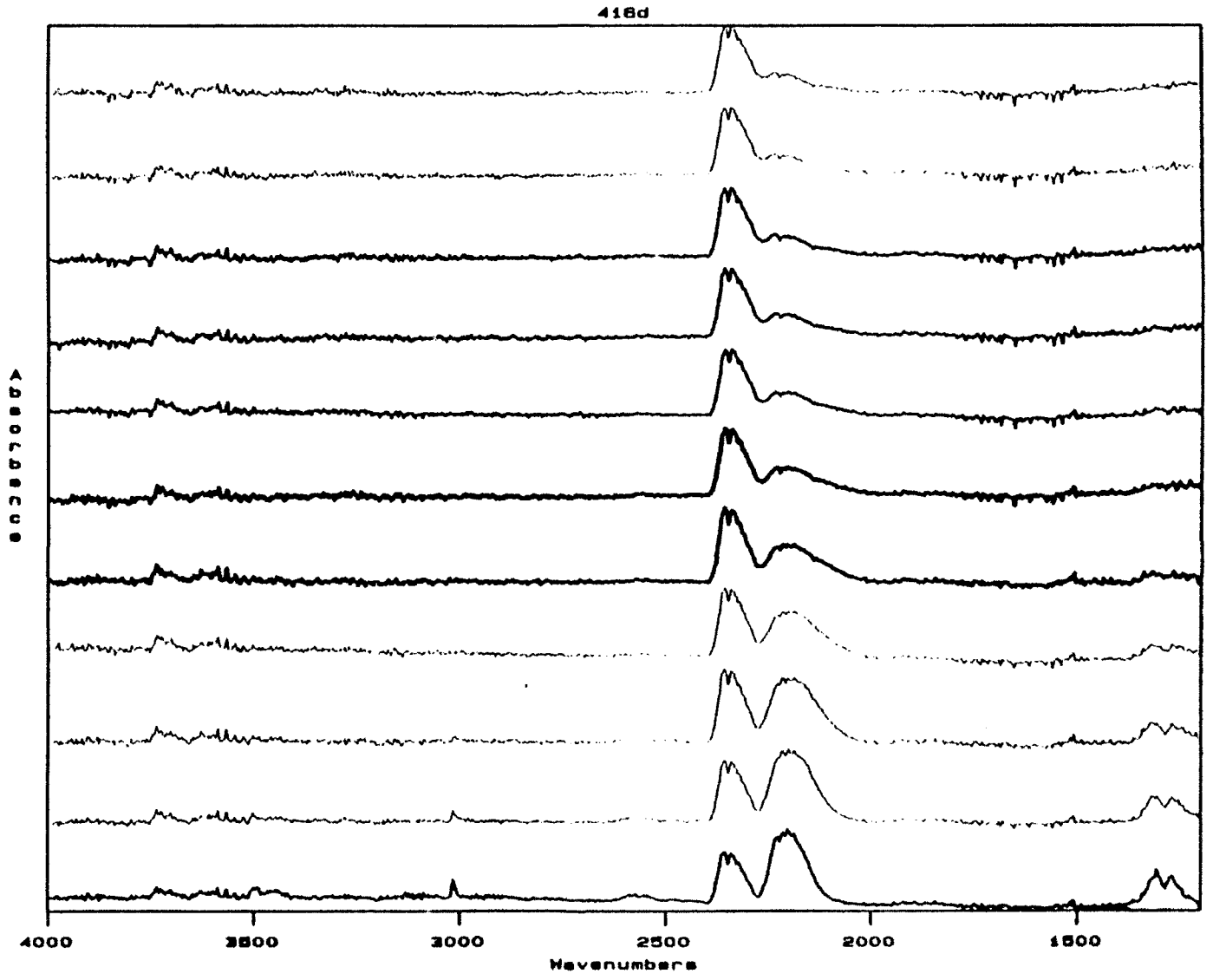

Figure 3. Line-of-sight absorbance spectra at lowest vertical position in the 17-torr CH_4/N_2O flame, with the line-of-sight path going from the center (bottom spectrum) to the shroud (edge) region (top spectrum) of the flame.

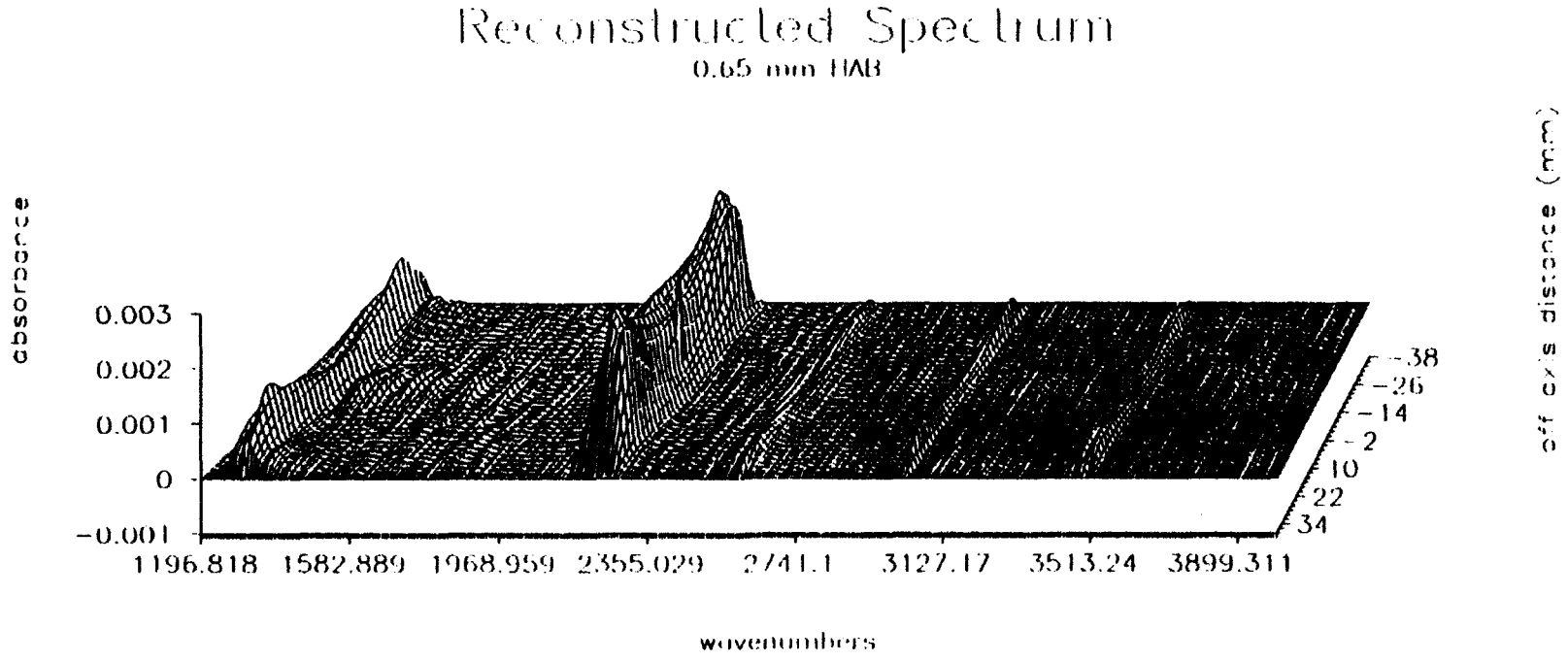

Figure 4. Tomographic reconstruction of localized absorbance (frequency vs. absorbance as a function of axial distance from the burner center) obtained from inversion of the spectra in Figure 3.

asymmetric and symmetric stretches of N_2O (at 2,223 cm^{-1} and 1,285 cm^{-1}, respectively) and by the CH stretch of methane (Herzberg 1950) (3,020 cm^{-1}). This figure shows that just above the burner surface there is a higher concentration (absorption coefficient times pressure) of methane and nitrous oxide at the edge of the burner than at the center of the burner. The luminous portion of the flame may affect the density of the fuel-oxidizer mixture in the nonluminous region by creating a temperature gradient within the nonluminous zone. The curvature of the luminous zone may be responsible for the varying concentration profile of methane and nitrous across the face of the burner at levels immediately above the burner surface. The spike in the reconstruction around 2,360 cm^{-1} is probably spurious and due to noise in the line-of-sight data. The error in all reconstructions, based on reconstructions of synthetic data (round off error used to simulate noise [Hughey and Santavicca 1982]), is believed to be approximately 10%.

Figure 5 shows relative peak absorbances (at one selected frequency for each species) for N_2O, CH_4, CO_2, CO, and NO as a function of distance from the burner center through a "slice" of the flame 0.65 mm above the burner surface. The actual spatial resolution can be no better than the diameter of the probe beam, in this case equal to 1.3 mm. Figures 6 through 10 show relative peak absorbances (at one selected frequency for each species) for N_2O, CH_4, CO_2, CO, and NO, respectively, at HAB surface from 0.65 mm to 13.05 mm as a function of distance from the burner center. The contour at the top of each figure is due to absorbance nearest the burner surface. Figures 9 and 10 show that for this flame, the region of highest CO and NO concentration, respectively, occurs approximately 3 mm above the burner surface in the nonluminous region of the flame. These results are in agreement with recently reported results on CH_4/N_2O low-pressure flames obtained using mass spectrometry (Vandooren, Branch, and Van Tiggelen 1992), although the peak in NO concentration followed by a decrease in the burned gas region has not been reported previously.

Obtaining absolute species concentrations requires knowledge of local temperatures and the dependence of the absorption coefficient for each species upon temperature (Ouyang and Varghese 1990). Work is presently being performed in which tomographic reconstruction of infrared diode laser spectra of CO will be used to obtain local temperatures.

5. CONCLUSION

We have shown that tomographic reconstruction using line-of-sight transmission spectra of low-pressure flames can be of use in evaluating the chemistry occurring within low pressure flames. At

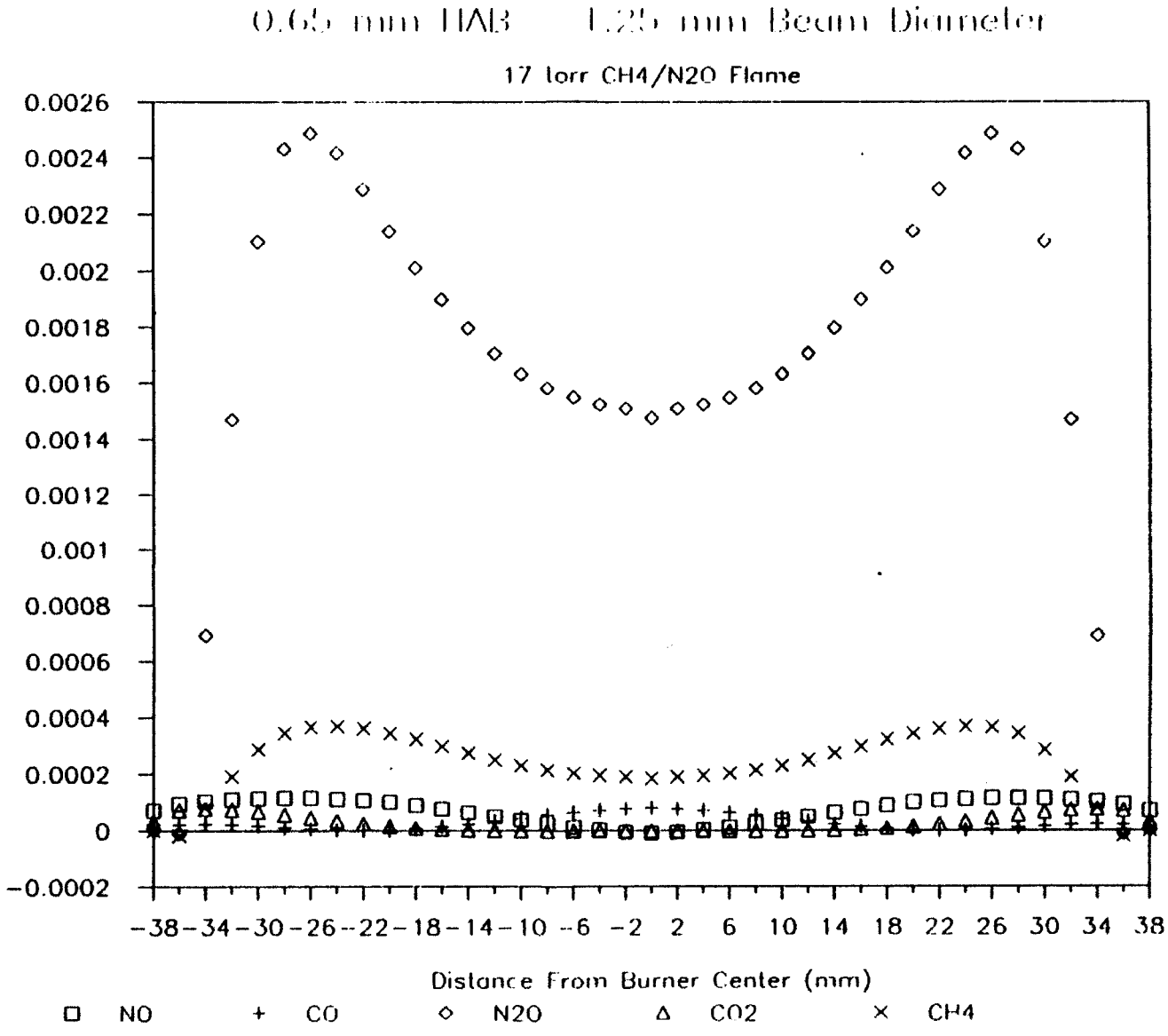

Figure 5. Relative peak absorbances for NO, CO, N_2O, CO_2, and CH_4 as a function of axial distance "r" from flame center 0.65 mm above the burner surface.

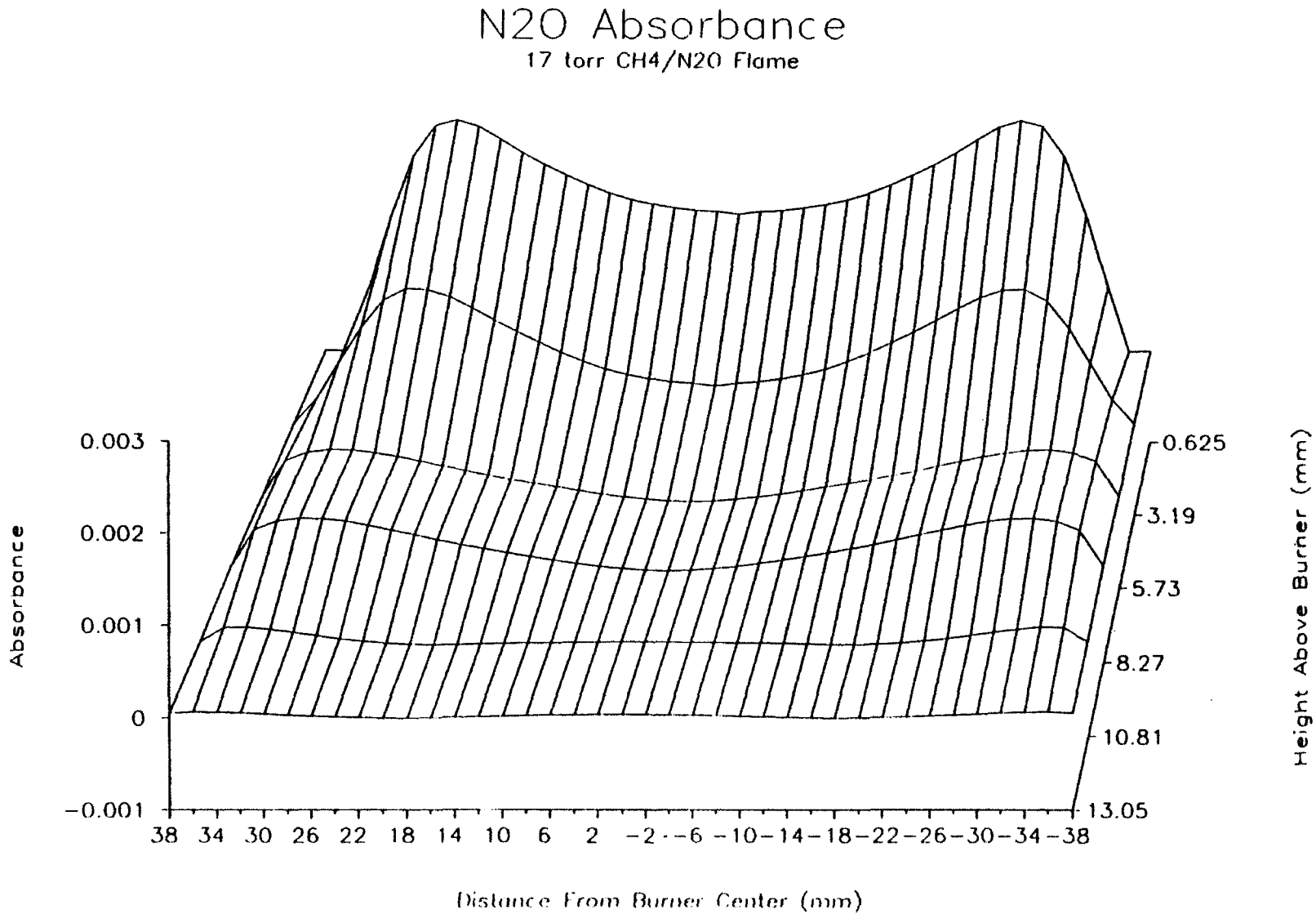

Figure 6. Relative peak absorbance of N_2O as a function of axial distance from flame center and HAB surface. Contour at top of figure is closest to burner surface.

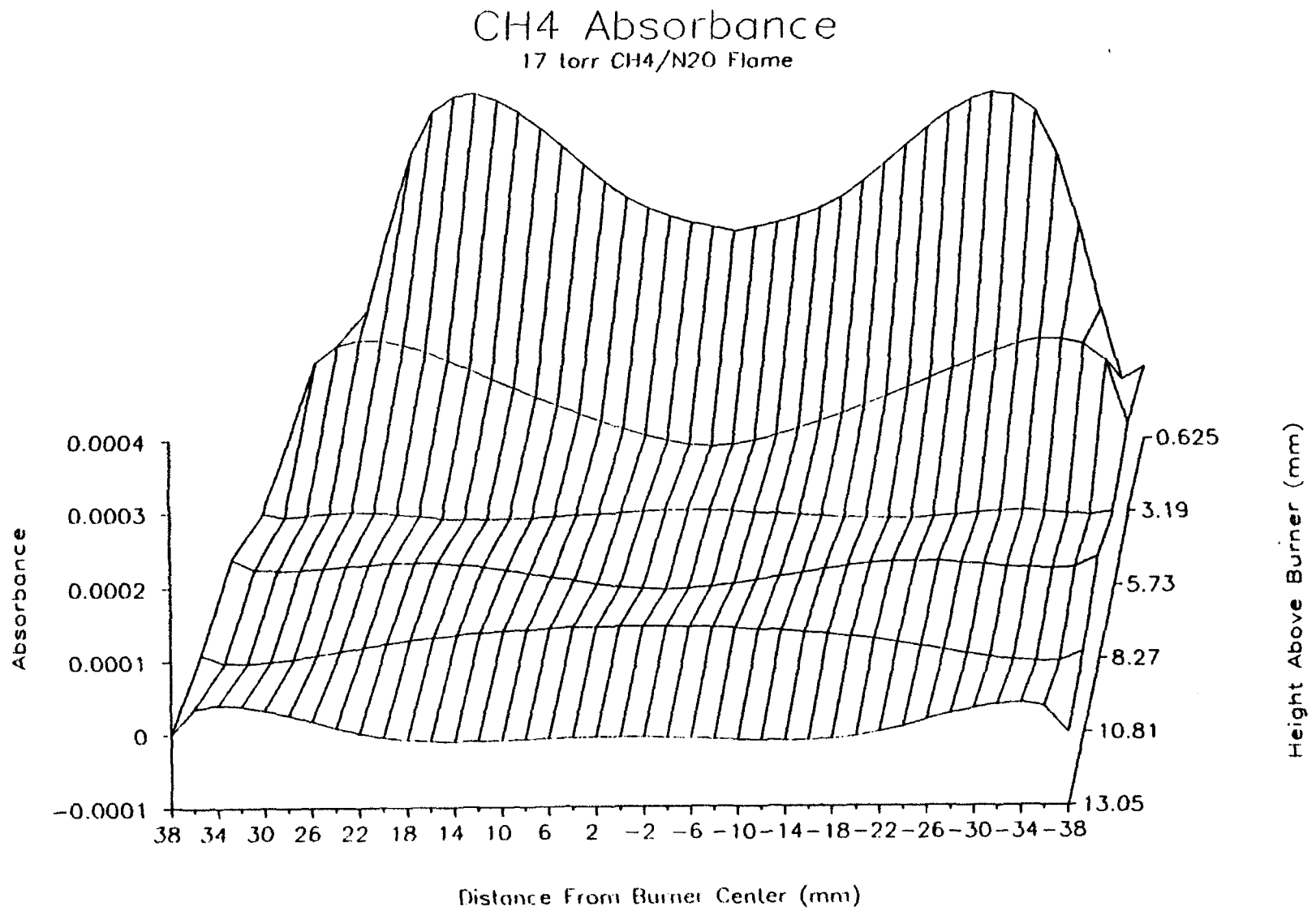

Figure 7. Relative peak absorbance of CH_4 as a function of axial distance from flame center and HAB surface. Contour at top of figure is closest to burner surface.

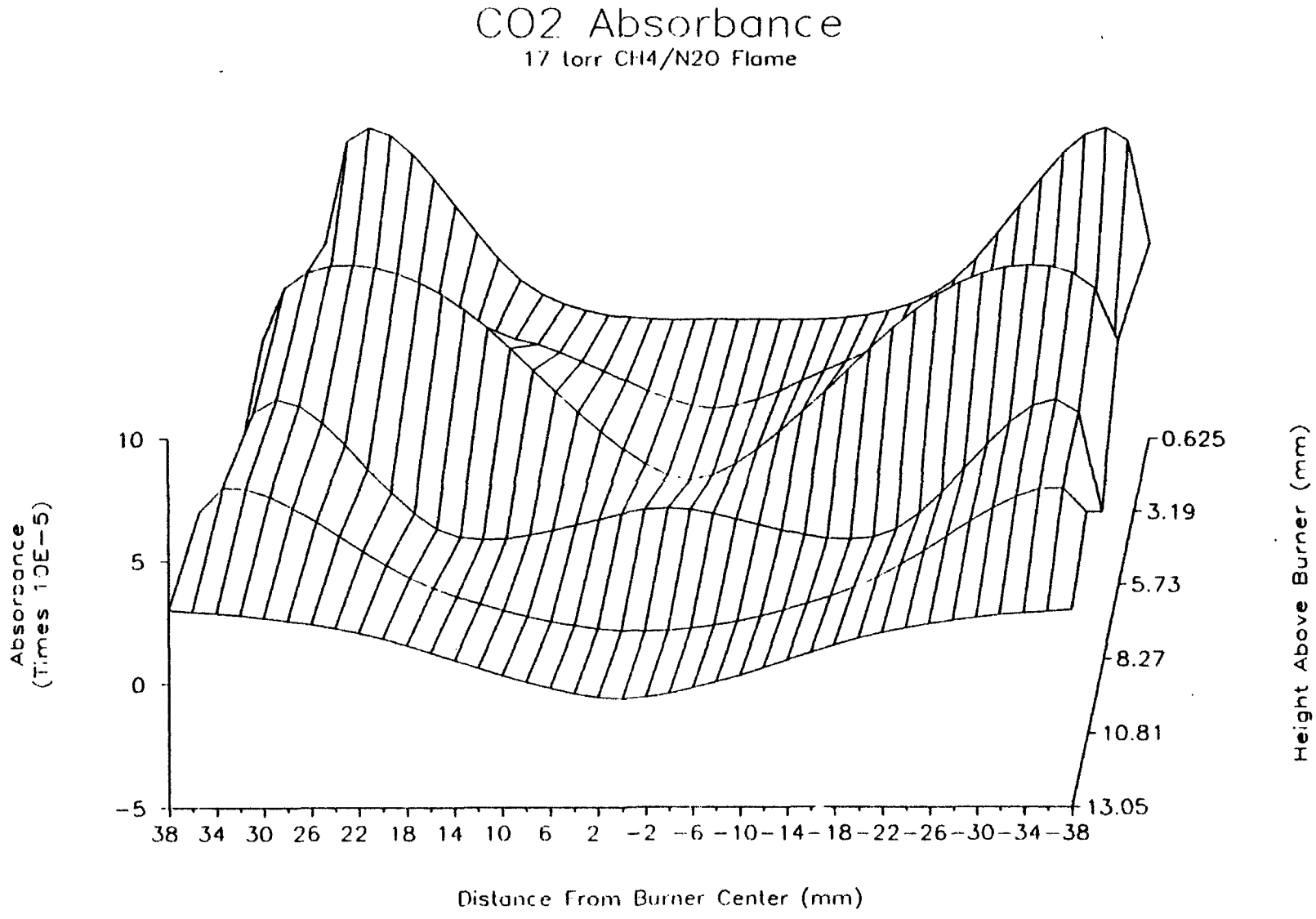

Figure 8. Relative peak absorbance of CO_2 as a function of axial distance from flame center and HAB surface. Contour at top of figure is closest to burner surface.

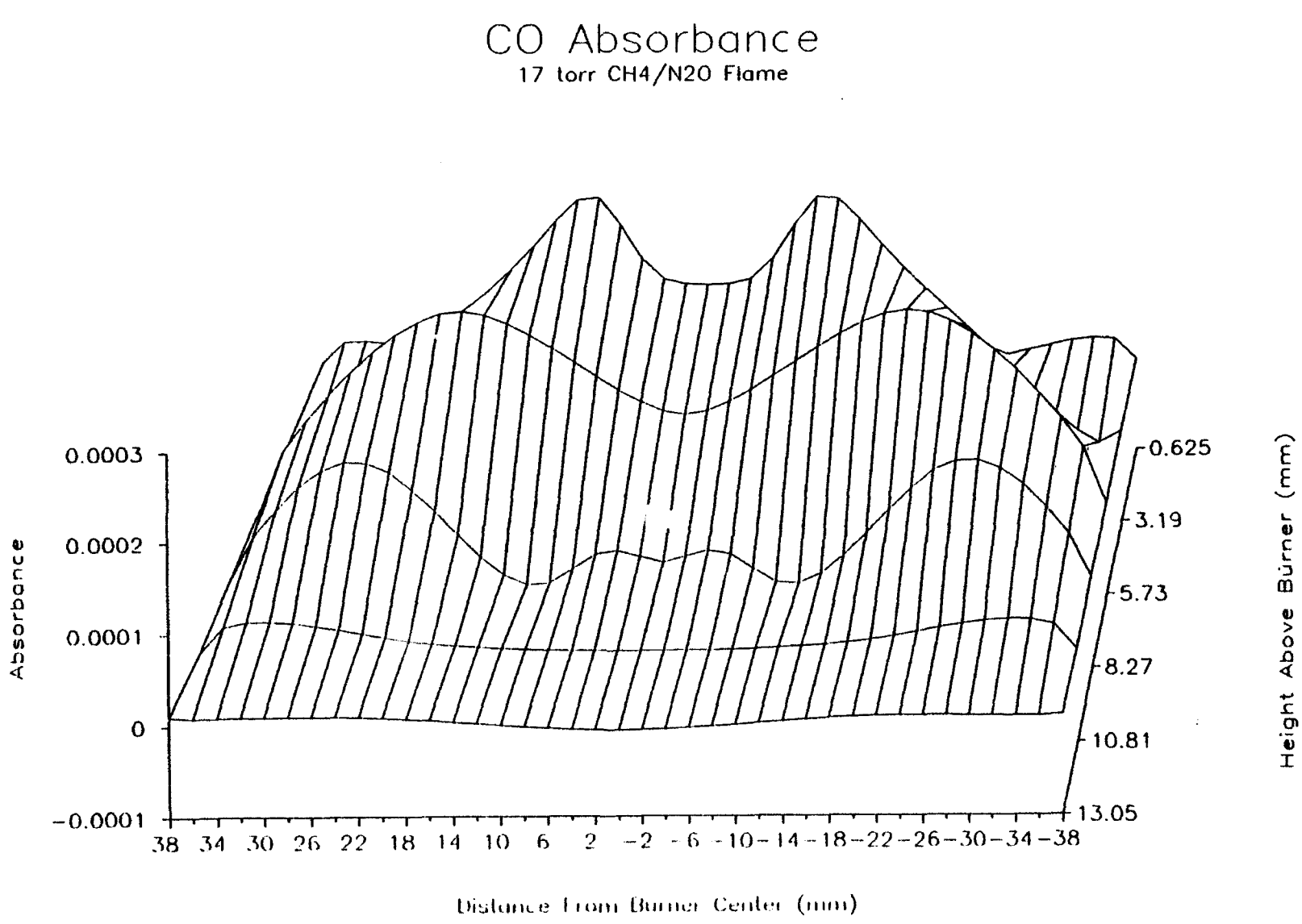

Figure 9. Relative peak absorbance of CO as a function of axial distance from flame center and HAB surface. Contour at top of figure is closest to burner surface.

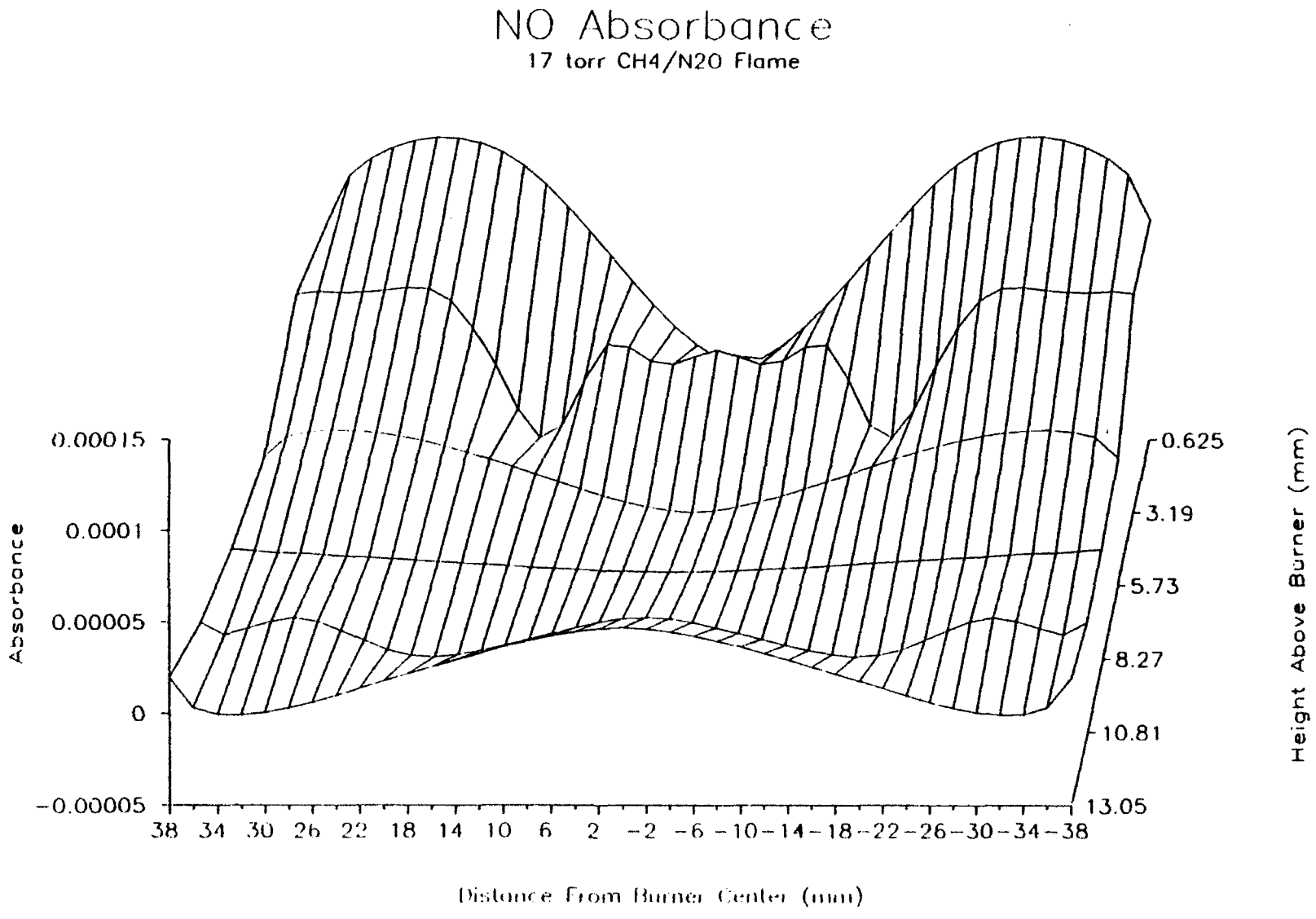

Figure 10. Relative peak absorbance of NO as a function of axial distance from flame center and HAB surface. Contour at top of figure is closest to burner surface.

present the technique is labor intensive, but with improved optics and array detectors, and improved signal-to-noise ratio, the technique may become routine. We are presently working on obtaining quantitative results by extending the technique to infrared diode laser spectroscopy, and using the information obtained as a calibration for tomographic reconstruction using FTIR line-of-sight spectra.

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